Electronic Supplementary Information for

Graphene Quantum Dots as Hole Transport Layers for High-Performance Organic Solar Cells

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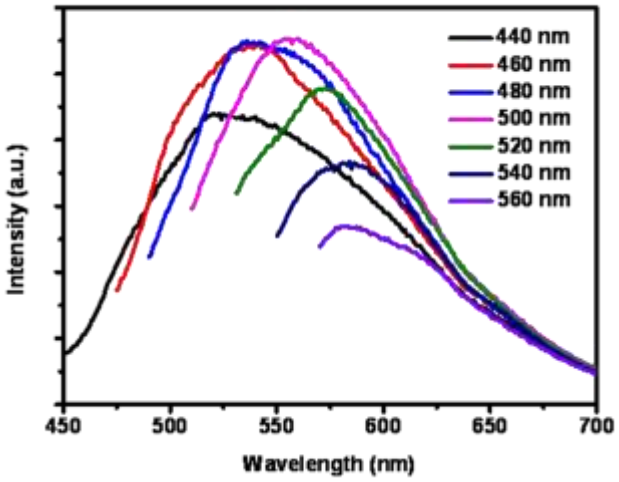


Fig. S1 PL spectra of GQDs with different excitation wavelengths increasing from 440 nm to 540 nm.

Figure S1 shows the photoluminescence (PL) spectra of the GQDs solution. The strongest emission peak at 556 nm is observed when the sample is excited under 500 nm. The emission wavelengths of the GQDs aqueous suspension are dependent on the excitation wavelengths. The PL peaks shift from 520 nm to 580 nm as the excitation wavelengths vary from 440 nm to 550 nm.

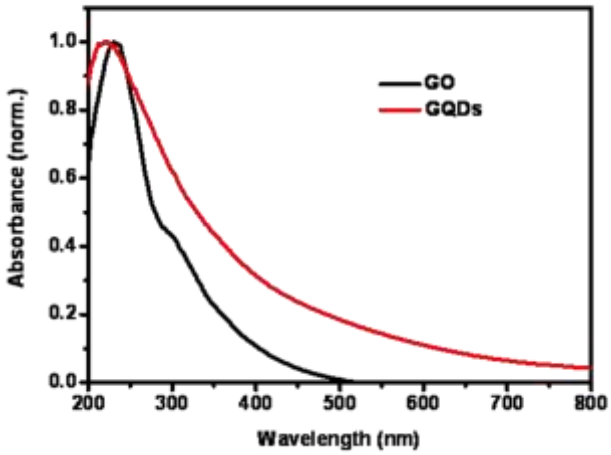


Fig. S2 UV/Vis absorption spectra of GO and GQDs aqueous solution.

The absorption peaks of GO and GQDs are 230 and 223 nm, respectively, indicating a

hypochromatic shift of the absorption peaks because of GQDs with less π-conjugation system.

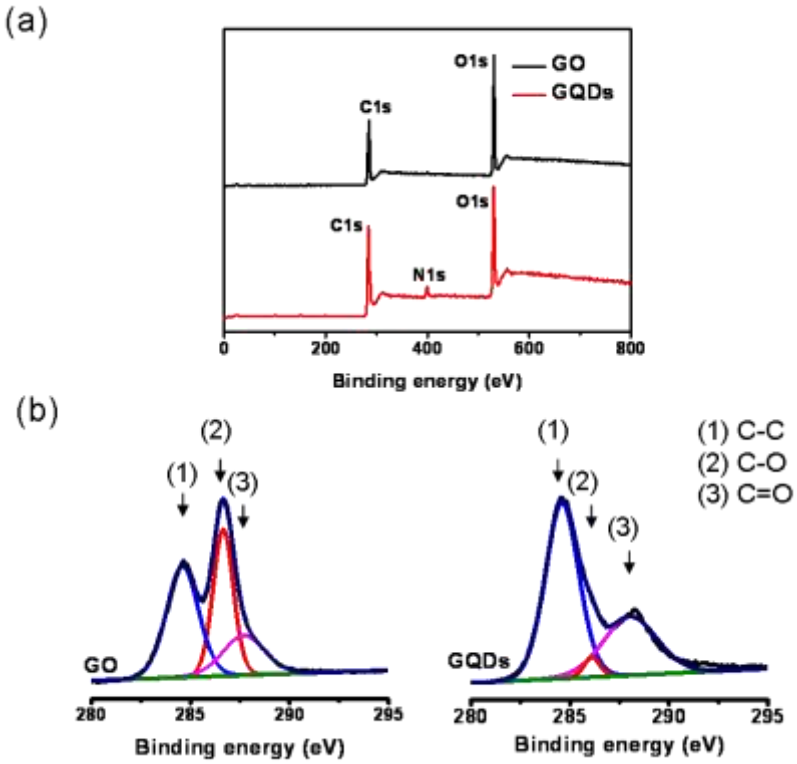


Fig. S3 (a) XPS survey spectra and (b) high-resolution XPS C1s spectra of GO and GQDs.

In high-resolution XPS C1s spectra of GQDs and GO, the presence of C–C (~284.6 eV), C–O (hydroxyl and epoxy, ~286.3 eV), C=O (carbonyl, ~287.8 eV) species were observed. The sum of C-O and C=O species in GQDs is lower than that of GO as shown in Figure S3(a). As shown in the XPS survey spectra, the less oxygen content of GQDs than that of GO indicates that GQDs possess less degree of oxidation than GO, due to the partial reduction of GQDs.

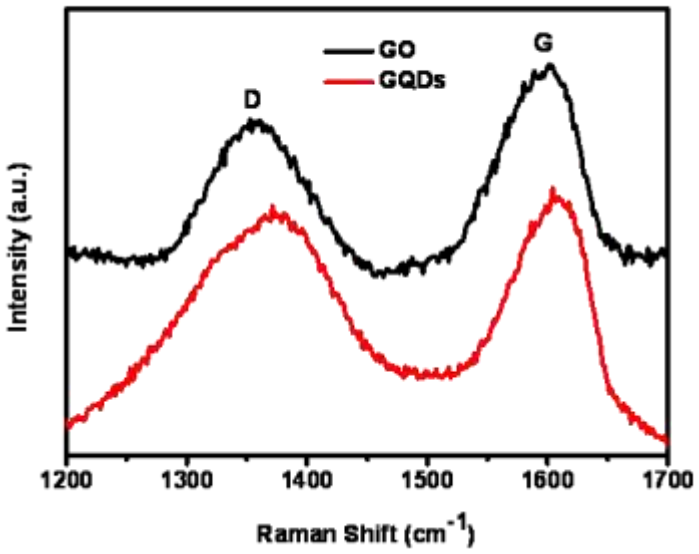


Fig. S4 Raman spectra of GO and GQDs.

The Raman spectra (Figure S4) of GO and GQDs show that the intensity ratio of the D-band to G-band (I_D/I_G) for GQDs (I_D/I_G = 0.94) is higher than that for GO (I_D/I_G =0.75).

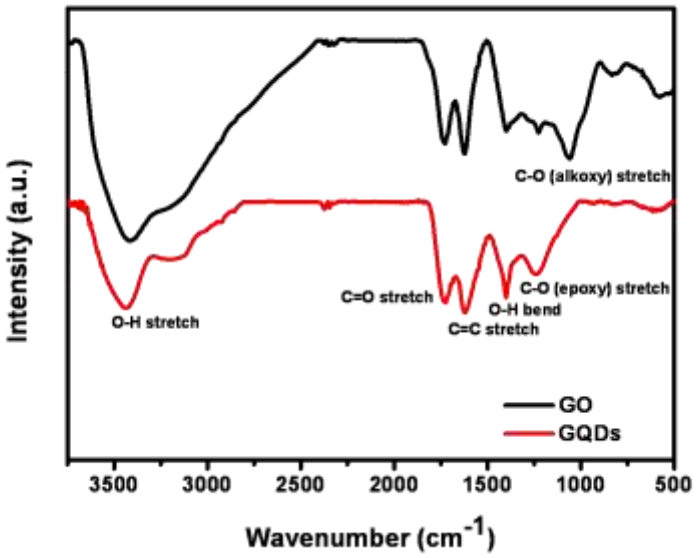


Fig. S5 FT-IR spectra of GO and GQDs.

The chemical functional groups on graphene sheets were characterized by FT-IR spectroscopy. As shown in Figure S5, the FT-IR peaks are corresponding to the oxygen functionalities, such as the C=O stretching vibration peak at 1726 cm^{-1}, the vibration and deformation peaks of O–H groups at 3410 cm^{-1} and 1400 cm^{-1}, respectively, the C–O (epoxy)

stretching vibration peak at 1226 cm^{-1}, and the C–O (alkoxy) stretching peak at 1057 cm^{-1}.

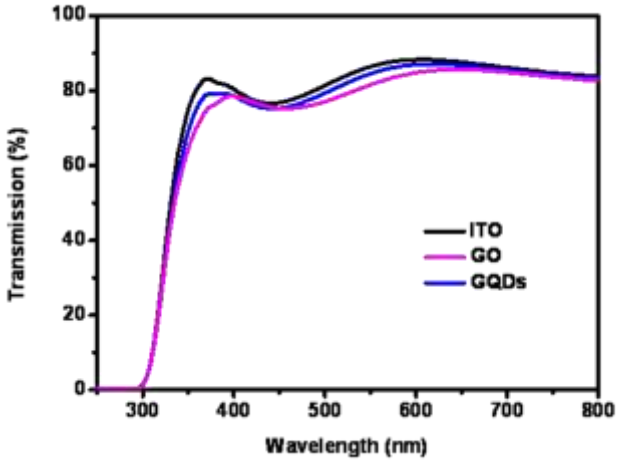


Fig. S6 Optical transmission spectra of GO (1.5-2 nm) and GQDs (1.5-2 nm) films deposited on ITO/glass substrates.

The optical transmission spectra of the ITO substrates which were coated with 1.5-2 nm GQDs or GO films were shown in Figure S6. The transparency of ITO doesn't significantly change with GQDs films and the transmittance of GQDs films is slightly higher than that of GO.

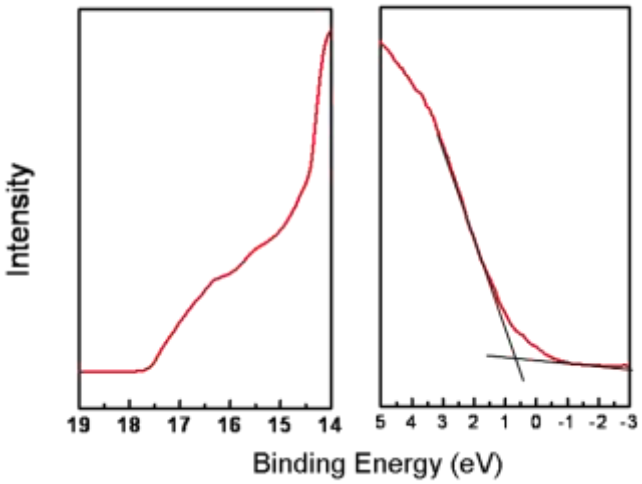
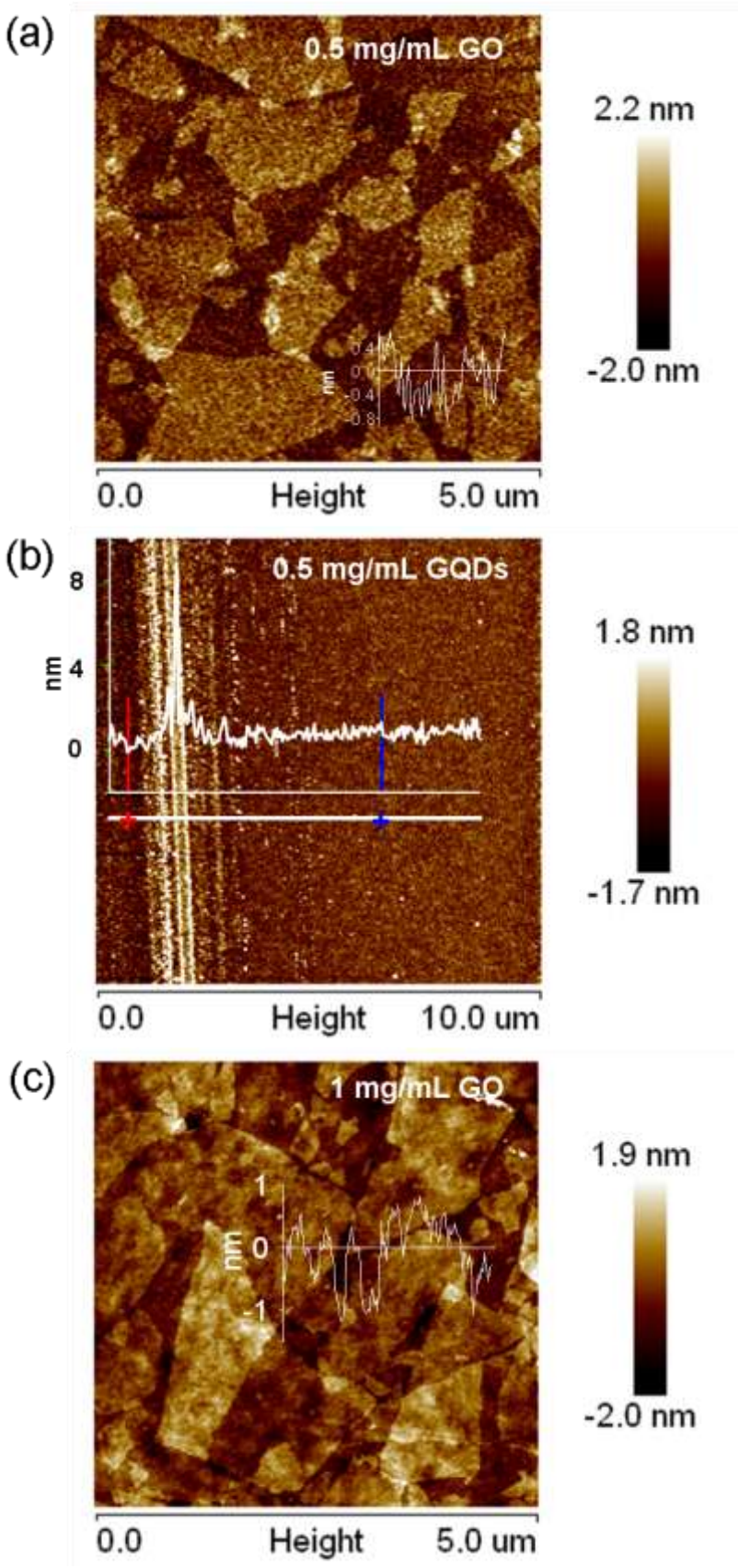
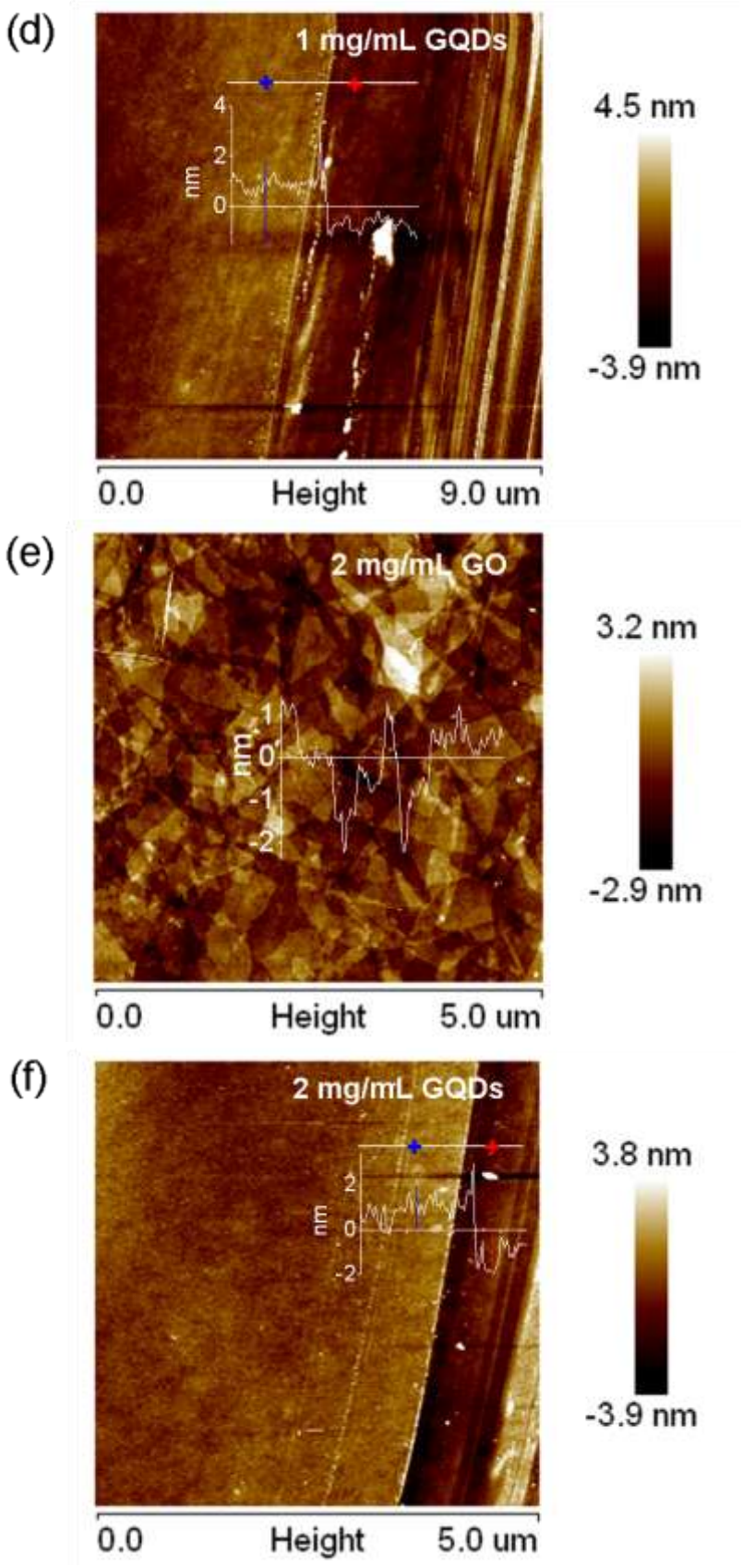
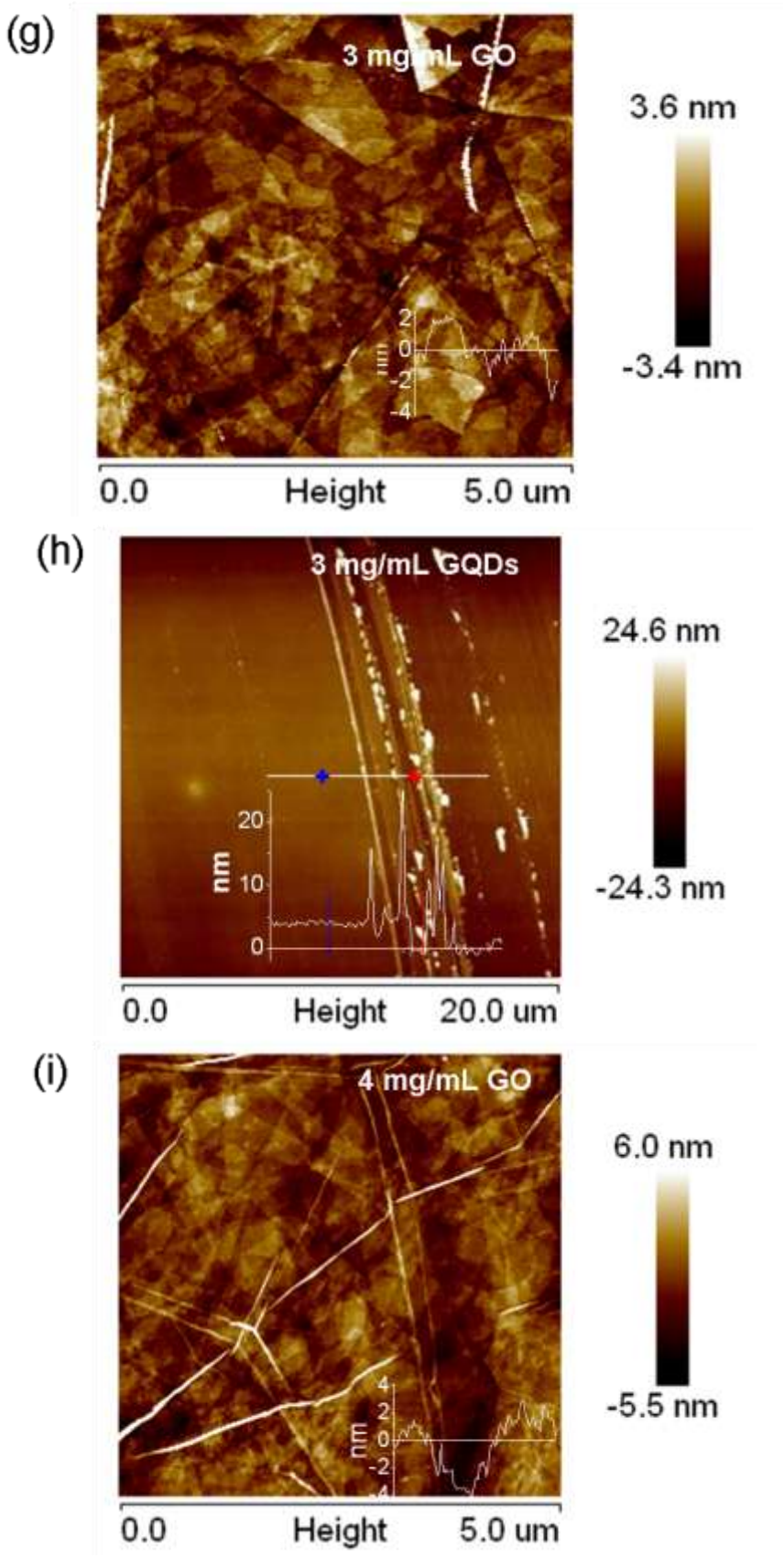


Fig. S7 Ultraviolet Photoelectron Spectroscopy (UPS) plots for GQDs.


(a)
0.5 mg/mL GO
2.2 nm
-2.0 nm
0.0
Height
5.0 um
(b)
0.5 mg/mL GQDs
8
nm
4
0
1.8 nm
-1.7 nm
0.0
Height
10.0 um
(c)
1 mg/mL GQ
1
nm
0
-1
1.9 nm
-2.0 nm
0.0
Height
5.0 um


(d)
1 mg/mL GQDs
4
2
0
nm
4.5 nm
-3.9 nm
0.0
Height
9.0 um
(e)
2 mg/mL GO
1
0
-1
-2
nm
3.2 nm
-2.9 nm
0.0
Height
5.0 um
(f)
2 mg/mL GQDs
2
0
-2
nm
3.8 nm
-3.9 nm
0.0
Height
5.0 um


(g)
3 mg/mL GO
3.6 nm
-3.4 nm
0.0
Height
5.0 um
(h)
3 mg/mL GQDs
24.6 nm
-24.3 nm
0.0
Height
20.0 um
(i)
4 mg/mL GO
6.0 nm
-5.5 nm
0.0
Height
5.0 um

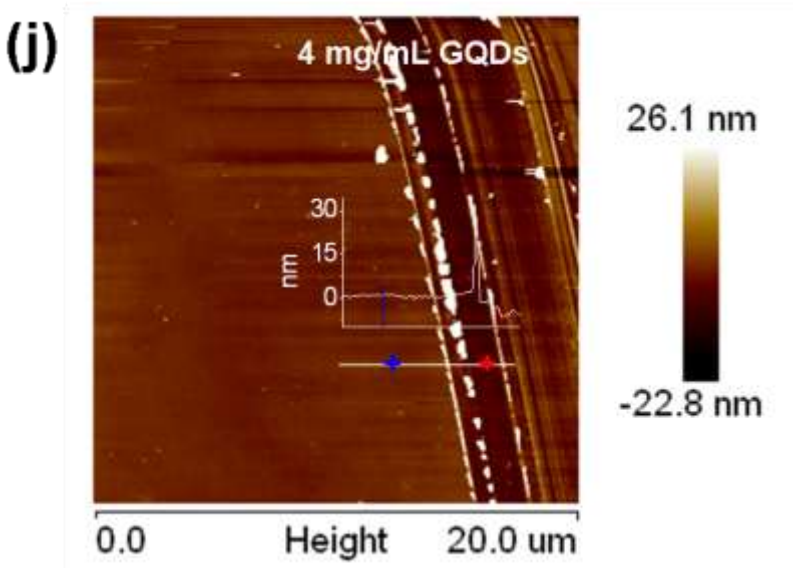


Fig. S8 AFM height images of GO and GQDs with different thickness deposited on SiO_2 /Si substrates. 0.5, 1, 2, 3, and 4 mg/mL of graphene-based films yield 1, 1.5-2, 3, 5, and 7 nm respectively. The sheets of GQDs are small and homogeneous, resulting in covering the substrate completely. Therefore, the thickness of GQDs films was measured though erasing a part of the films.